

# Coal-based Clean Energy Production

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## Abstract

The results of the “energy production” division of a 4-year development work of an eco-city project at Lanfang, China, are reported. A novel “coal to methane” process has been developed from laboratory scales into a pilot plant, where the coal was treated through a “catalytic supercritical water gasification” process. The overall efficiency achieved in this pilot plant is as high as 75% with thermal energy recovery, compared to the 56% efficiency of “coal to methane” processes using existing gasification technologies. The sequestered carbon dioxide (CO<sub>2</sub>) from the production (gasification) process is further used for microalgae growth, which is then refined into biodiesel and other value-added by-products. The whole integrated “energy production” system can achieve low emissions of CO<sub>2</sub> and other contaminants.

## Keywords

*Coal Gasification; Supercritical Water; Microalgae*

## Introduction

Coal continues to play a vital role in energy supplies, especially in some developing countries. Besides the pollution from acid gas (SO<sub>2</sub>), mercury and particulate matters, the inevitable CO<sub>2</sub> emission from using carbonaceous fossil fuel sources is one of the most significant contributors to the greenhouse effect and global warming.

Among the fossil fuels, natural gas is one of the most environmental-friendly energy sources with low CO<sub>2</sub> and other pollutant emissions. Table 1 shows the carbon to hydrogen ratios in the most commonly used fuels, which implies the relative strength of carbon emissions. This demonstrates that before “hydrogen economy”, natural gas is the cleanest fuel type among these fossil fuels. Recent development of shale gas exploration did positively contribute to solving world energy crisis with reduced CO<sub>2</sub> emission. However, conversion of coal into natural gas (CH<sub>4</sub>) is required for some regions and several “coal to methane” projects have been recently approved and under construction (Kopyscinski, 2010). However, the current gasification technologies are not intended for

natural gas production. Normally, they are developed for syngas production and further for fertilizers and other chemical syntheses. If they are adopted for methane production, a series of treatment, such as water shift and mathanation etc., is necessary. In North Dakota, USA, there has the first coal to methane project in the world (Dakota Gasification Company, 2006), where Lurgi gasifiers (Esenlohr, 1974) are utilized with a series of downstream syngas treatment, mathanation process, which affected the production costs and the overall energy efficiency of the plant. More efficient coal to methane technology is urgently required in the market.

TABLE 1 FUEL TYPES AND THEIR CARBON TO HYDROGEN RATIOS

Era	Steam engine	Internal combustion engine	Gas turbine	Fuel cell
Fuel	Coal	Oil	Natural gas	Hydrogen
Energy state	Solid	liquid	Gas	Gas
C:H	7~12:1	0.45~0.48:1	0.25:1	0

Gasification involves complicated chemical reactions which consist of oxidation, and secondary water shift and possible mathanation, although the mathanation can only happen at a temperature lower than ~700°C. For methane production from coals (e.g., bitumen, lignite, and peat), higher pressure and lower temperature are beneficial, as mathation reaction reduces molecule number and is endothermic. Supercritical water (Temperature>374°C and Pressure>22.1MPa) (Kruse, 2009) would provide a possible environment. Therefore, gasification of coals in supercritical water is proposed for this research. With a favorable reaction condition, the process will reduce or eliminate the needs for sub-processes.

Improved system efficiency in the production processes will reduce CO<sub>2</sub> emission accordingly. By elevating the gasification pressure and reducing its temperature, the system energy efficiency can be improved. The waste heat recovery in existing gasification technologies is a difficult task, because of the poor heat transfer coefficient of the gaseous products. While with supercritical water gasification,

the pressure reaches 23.0 MPa or higher, which is realized by pumping liquid and solid phase (coal-water slurries). After reaction, the "waste" heat at 500~700°C carried by high pressure gaseous products will be recovered through a turbine to generate power, though there are still engineering challenges for turbine to work with this gas stream. This "supercritical water gasification" process could include an internal "Rankine power generation cycle". The improved efficiency will result in lower CO<sub>2</sub> emission, based on unit energy produced. However, there is still some amount of CO<sub>2</sub> emission from the process.

For large power generation plants or fossil fuel related process industry, carbon capture and sequestration (CCS) has been proposed. Because of its uncertainty and high additional cost, CCS technologies are considered as short-term solutions, as there are still concerns about the environmental sustainability of these processes.

Alternatively, a promising technology is the biological capture of CO<sub>2</sub> using microalgae (Chisti, 2011). These microorganisms can absorb CO<sub>2</sub> using solar energy with efficiency tens times greater than terrestrial plants. Moreover, the capture process using microalgae has the following advantages: (i) environmentally sustainable; (ii) absorbing solar energy; and (iii) co-producing high value added materials based on biomass, such as human nutrition food, cosmetics, medical drugs, fertilizers, biomolecules for specific applications and most important – (iv) biofuels.

This work outlines a clean energy solution, which combines a novel gasification technology and CO<sub>2</sub> capture with microalgae.

### Supercritical Water Lignite Gasification

Coal gasification technology has been already widely used in industries such as in fertilizer, methanol and other chemicals production. These gasification technologies are normally at temperatures as high as >1000°C, such as Lurgi gasifier, Texaco gasifier and Shell gasifier. All of these approaches involve the gasification of coal with steam and oxygen. Differences are related to gasifier configuration, operating parameters and ability to handle different types of coals.

Lignite, as a low-rank of coal, is a nonhomogeneous solid material which contains a significant mineral matter, largely silicon, aluminum, magnesium, and iron. The mineral compositions vary dramatically. In general, it has complicated, coupled and interacted

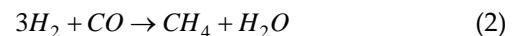
chemical reactions when it is gasified with steam at high temperatures 550~2000°C, and a recent gasifier under development by RocketDyn (Pratt-Whitney) has 2700°C gasification temperature.

The dominant gasification reaction can be regarded as:



Where coal is assumed as pure carbon and this reaction requires heating at 600°C and 25 MPa.

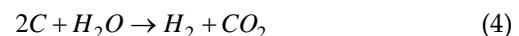
Methanation occurs with hydrogen and carbon monoxide, and release heat.



However, the methanation requires a different CO and H<sub>2</sub> proportion. An intermediate step called the "water gas shift" is required:



The overall reaction is:



It requires heat input for this overall reaction. This heat is normally supplied by oxidation of coal:

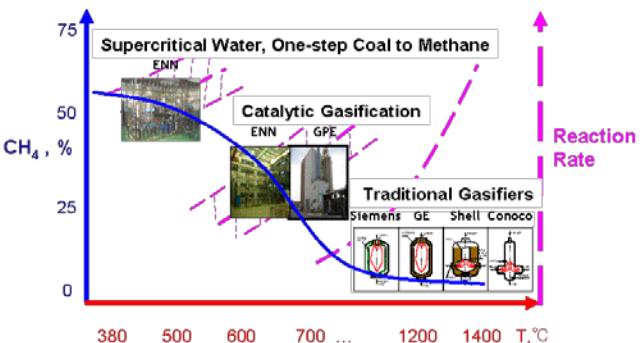
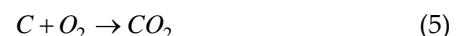


FIG. 1 GASIFICATION THERMODYNAMICS AND KINETICS

(The technology sources: ENN-ENN Group (China), GPE-GreatPoint Energy Inc. (USA), Siemens (Germany), GE-General Electronics (USA), Shell (Great Britain), Conoco-Conoco-Philippines (USA). Traditional gasifiers are operating at 1200~1400°C, catalytic gasification at 700~800°C, and the supercritical water catalytic coal gasification is expected to operate at 500~700°C.)

The reaction (4) to form CH<sub>4</sub> is not obvious at high temperature, as it is very strong endothermic reaction. High temperature is necessary for a fast chemical kinetics, so that scaling-up for mass production can be realized with a reasonable reactor size. To increase the reaction activity at lower temperature, catalyst is an option. Therefore, catalytic coal gasification at 700~800°C is developed (Hirsch, 1982; Nahas, 1983). Following this trend, Figure 1, it could be concluded that at even lower temperature, a better methane yield ratio can be reached (Yamaguchi, 2009; Zhang, 2010; Vostrikov, 2011). To keep the gasification process with

high reaction activity, a novel catalysis reaction in a supercritical water environment is proposed in this work with additional thermal energy recovery.

Supercritical water gasification (SCWG) includes hydrothermal conversion, which utilizes water above its critical point (374°C, 22.4 MPa).

Simulation using ASPEN shows that an almost total conversion of coal to CH<sub>4</sub> with very high yield concentration is possible, see Figure 2.

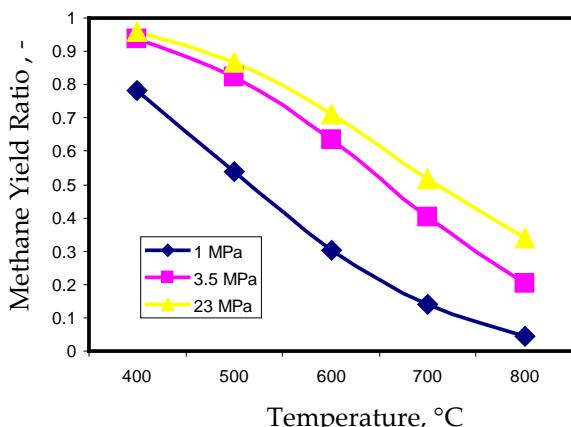


FIG. 2 ASPEN SIMULATION RESULTS OF COAL GASIFICATION AT VARIOUS TEMPERATURE AND PRESSURE

This equilibrium simulation doesn't show the kinetics of the reactions. Experiments and pilot-plant data identified that at supercritical water conditions an acceptable reaction rate is achieved, especially with catalyst. Nevertheless, temperature effect on kinetics is always significant.

Therefore, this SCWG offers several substantial benefits:

- 1) high surface area after SCW extraction of coal, similar to activated carbon;
- 2) high catalyst surface area due to transcritical catalyst dispersion;
- 3) high methane yield, so that the methanation process can be reduced or eliminated;
- 4) better heat transfer properties of the effluent, so that higher energy recovery rate is possible;
- 5) power generation from the high pressure products is possible;
- 6) CO<sub>2</sub> can be captured and separated from high pressure products in liquid phase.

## Experimental

Lignites (Yamaguchi, 2009; Zhang, 2010; Vostrikov, 2011) are preferred feedstocks for SCWG, as their high moisture content makes their utilisation in power plant inefficient. SCWG uses wet feedstocks in slurry

form and the water participates in the chemical reactions (Kruse, 2008). Sewage sludge as municipal and industrial wastes was treated using SCW. In this research, from high organic concentration waste water, sewage sludge, biomass (including microalgae), peat, to lignite, a large variety of feedstock has been studied in the laboratory bench scale units and tested in the pilot-plant. A lignite sample from South China used intensively in this study has the analysis data shown in Table 2.

TABLE 2 ANALYSIS DATA OF A LIGNITE SAMPLE, %

C	H	O	N	S	Moisture	Ash
30.10	2.42	10.04	0.63	1.17	44.75	10.89

The density, solubility and transport parameters of water will increase with increased pressure, therefore, enhancing the kinetics of lignite gasification reactions. Moreover, supercritical water will participate in the chemical reactions, hydrothermally or by direct hydrolysis. A higher lignite conversion rate can be achieved when compared to pyrolysis with absence of water, Figure 3.

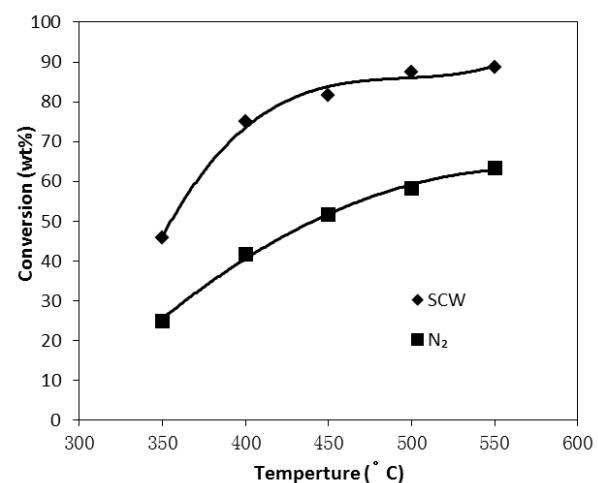


FIG. 3 COMPARISON OF CARBON CONVERSION RATE OF LIGNITE IN SUPERCRITICAL WATER AND IN INERT ENVIRONMENT

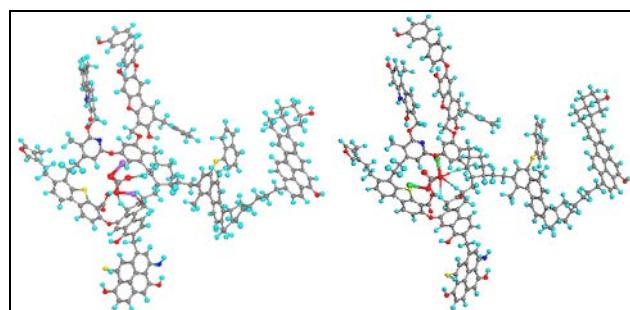


FIG. 4 ADSORPTION OF Na<sub>2</sub>CO<sub>3</sub> AND K<sub>2</sub>CO<sub>3</sub> ON COAL MOLECULES (LEFT - Na<sub>2</sub>CO<sub>3</sub>, RIGHT - K<sub>2</sub>CO<sub>3</sub>)

Furthermore, catalysts would also improve the reaction kinetics at low temperature. It has been

known that alkali metal salts of weak acids, such as  $K_2CO_3$ ,  $Na_2CO_3$ ,  $K_2S$  and  $Na_2S$ , can catalyze the steam gasification of coal (Hirsch, 1982). Figure 4 revealed the adsorption of the salts  $K_2CO_3$  and  $Na_2CO_3$  on coal molecules, through a molecular simulation by this work.

From the molecular simulation results, a stronger bond between coal and  $Na_2CO_3$  is found relative to  $K_2CO_3$ . Experimental results identified that a better  $CH_4$  yield is achieved with  $Na_2CO_3$  as catalyst (Zhang, 2012). However, some other literatures showed a reversed result that  $K_2CO_3$  is a better catalyst over  $Na_2CO_3$ . This new result is encouraging, as  $Na_2CO_3$  is a cost effective alternative to  $K_2CO_3$ . The optimal concentration of catalyst is confirmed to be 10–20% with the South China lignite, which showed very close results to previous researches (Hirsch, 1982).

A pilot-plant is built to test the process and equipment (reactors, heat exchangers, and separators etc.) integrity, and to find and solve engineering problems, such as multiphase flow instability, erosion, corrosion and clogging etc.

The first step of the process is a hydrothermal extraction at supercritical condition, where volatile substances are extracted and separated from the coal particles at sub- and supercritical water environment. Hydrothermal reaction involving  $H_2O$  molecules occurs, together with pyrolysis. After this step, the coal particles become porous and posses large surface area.

With the addition of catalyst solution, the effluent from the extraction stage is chilled to subcritical condition. The catalysts,  $K_2CO_3$  or  $Na_2CO_3$  or their mixtures, have a high solubility at subcritical water, and almost insoluble in supercritical water. After the catalyst mixer, the extractant phase is separated and recovered as liquid fuels or refined as fine chemical products. The resulted porous coal structure with high surface areas becomes highly active in the gasification process. This residual coal slurry with catalyst is then heated to supercritical state, where the catalyst will crystallize from the mixture and form fine particles due to the sudden solubility drop. The particles, sized in a range of 100~5000 microns, exhibit a very high catalytic activity.

Then, within the catalytic gasification process in supercritical water, the reaction happens with “activated” coal particles and highly active catalyst fine particles. The supercritical water itself also possesses a high reactivity than normal steam.

Therefore, the gasification temperature can be reduced down to 550~650°C with a good conversion rate. A 80% conversion rate for SCWG at 23 MPa and 600°C will take about 0.5 ~ 2.0 minute; if with 5%  $Na_2CO_3$  as a catalyst the conversion time will be reduced to 0.67 minutes. While, with the same lignite as feedstock, catalytic gasification at 700°C at 3 MPa will take about 6~10 hours to reach the same conversion ratio (Naha, 1983).

Within the reactor, there is a series of reactions where the exothermic heat of formation of methane is provided to gasify the coal and produce hydrogen and carbon monoxide, which are the more usual products of coal gasification. In the supercritical water environment, the reactor will need much less heat input (only ~ 7% of high temperature gasification), e.g., much less oxygen.

As an option for process integration, hydrogen is recycled back to the reactor after product separation. Finally,  $CH_4$  is obtained as the product (Table 3.)

TABLE 3 THE DRY GASIFICATION PRODUCT

Product	Vol%
$CH_4$	49.9
$H_2$	44.6
CO	4.0

### Integrated Coal Gasification and Microalgae Cultivation

Hydrogen is then sent back to the reactor where hydrogasification under supercritical water is carried out with high hydrogen concentration. Early work (Anthony, 1976) showed that with high pressure hydrogen, a substantial portion of the carbon in raw coal can be converted to methane more rapidly than the carbon in char. The Arizona Public Service (USA) (Cicero,2009) focuses on the so-called hydrogasification process in which coal will be gasified with hydrogen at moderate temperatures (870°C) and high pressures (7.0 MPa). The methane containing syngas is directly produced in the gasifier without any catalyst. Figure 5 showed the hydrogen cycling operation integrated with supercritical water gasification.

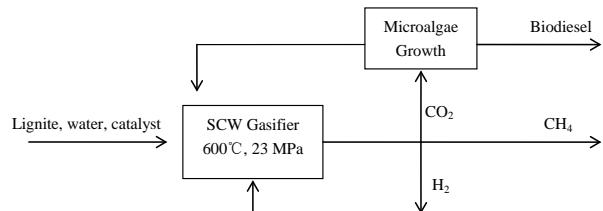


FIG. 5 COMBINED COAL GASIFICATION AND MICROALGAE GROWTH

A very small portion (<0.1%) of solar energy reaching Earth is absorbed by vegetation, forming the critical source of lives. For commercial feasibility, fast growing species are required, such as microalgae, which absorb about 100 times of CO<sub>2</sub> as forestation does and produce ~ 50 times of oil as soya bean. The concept of using algae to make fuels was already discussed 50 years ago, but a concerted effort began with the oil crisis in the 1970s. Large research programs in Japan and the United States focused on developing microalgal energy production systems. From 1978 to 1996, the U.S. Department of Energy's Office of Fuels Development funded a program to develop renewable transportation fuels from algae. The main focus of the program, known as the Aquatic Species Program (ASP), was the production of biodiesel from high-lipid-content algae grown in ponds, using waste CO<sub>2</sub> from coal-fired power plants. In Japan, the government financed a large research project entitled "Biological CO<sub>2</sub> Fixation and Utilization" from 1990 to 1999. These programs (Lam, 2012) generated meaningful results, such as promising lipid production strains, open production systems (raceway ponds), and principles for photobioreactor design which uses fiber optics to bring light inside the systems. However, economics on a large scale has not yet been proven (Wijffels, 2010; Singh, 2011). Since 2006, ENN focuses on the following aspects to improve the economy of microalgae "farm".

- 1) Advance technologies in three areas: genetic, bioreactor and process engineering;
- 2) By-produce value-added products, such as human nutrition additives, animal feeds, and fine chemicals;
- 3) Utilise gasification waste water for microalgae growth, as it contains nutritious ammonia and minerals;
- 4) Establish innovative commercialization pathways.

Algae is a very promising source of biomass in this context as it sequesters a significant quantity of carbon from atmosphere and industrial gases (mainly CO<sub>2</sub>) and is also very efficient in utilizing the nutrients from industrial effluents and municipal wastewater. Therefore cultivation of algal biomass provides dual benefit, and biomass for the production of biofuels and also protects our environment from air and water pollutions.

The results on microalgae from laboratory tests, pilot-scale experiments, a demonstration project in the eco-

city, to the commercialization project in a deserted field near a coal-to-methanol plant in Inner Mongolia (China), will be reported in a separate paper. For example, ENN51 microalga species can grow at environmental temperature of -10 ~ 4°C, with a yearly yield of 5.2 kg/m<sup>2</sup> dry alga powder (2.1 kg/m<sup>2</sup> oil, e.g. bio-crude).

#### Material/mass balance of the system:

For the gasification pilot-plant, the material balance is shown in the following Table 4, Table 5 and Table 6. The results are from the designed scale and calibrated from several experimental runs.

TABLE 4 INPUT MATERIAL BALANCE OF THE GASIFICATION PILOT-PLANT

Items	State	Quantity	Unit	Comments
Lignite	solid	720	kg/day	
Water	liquid	1680	kg/day	Slurry making
Oxygen	liquid	200	kg/day	
Natural gas	gas			Not measured, for start-up, against heat loss
Cooling water	liquid			Not measured, recycled

TABLE 5. OUTPUT MATERIAL BALANCE OF THE GASIFICATION PILOT-PLANT

Items	State	Quantity	Unit	Comments
CH <sub>4</sub>	Gas	76	kg/day	
H <sub>2</sub>	Gas	14	kg/day	vol% is 120% of CH <sub>4</sub>
CO	Gas	13	kg/day	recycled
CO <sub>2</sub>	Gas	535	kg/day	recycled
Others: C <sub>2</sub> H <sub>6</sub> , H <sub>2</sub> S, NH <sub>3</sub> , Ash, not continuously recorded				

In addition, solar energy is harvested in the form of biomass, specifically microalgae cultivation. A greenhouse for microalgae production is built with a area of 17600 m<sup>2</sup> (4.28 acres). Waste water containing NH<sub>3</sub> and minerals from the gasification section is pretreated and used for microalgae cultivation. The contained NH<sub>3</sub> serves as a fertilizer to the plants.

TABLE 6. LARGE SCALE MICROALGAE CULTIVATION

Items		Comments
Land use	17600 m <sup>2</sup>	
Algae produced	91 ton/year	dry powder
Biodiesel	36 ton/year	
CO <sub>2</sub> emission reduced	286 ton/year	

#### Future Trends and Perspectives

For the conversion from lignite to natural gas, the retention time plays an important role, as shown in Figure 6 in the collective results from ENN's pilot-plant and the data from real field operations.

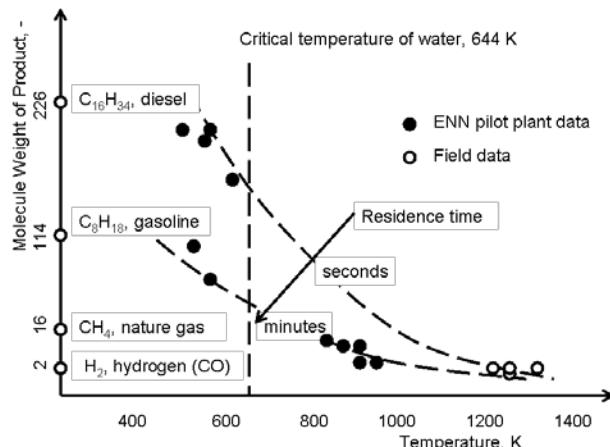


FIG. 6 HYDROTHERMAL TREATMENT OF LIGNITES

This may lead to new designs of kinetic dominant processes. With the tubular reactor design, the flow pattern and retention time, even a distributed temperature profile can be easily reached. However, more detailed test data for each different feedstocks are required, and engineering issues are also critical to solid contained systems.

## Conclusions

From the 4-year research and development on a coal-based low-carbon energy production project, where a low ranked coal-lignite is gasified in a supercritical water environment and the CO<sub>2</sub> generated is used for microalgae cultivation, a satisfactory result is achieved. The laboratory experiment, kinetic measurement, process demonstration and pilot-plant results have revealed a positive outcome, with which further engineering development is required. A 75% overall efficiency is achieved in the pilot plant. CO<sub>2</sub> from the gasification process is used in microalgae growth, which is then refined into biodiesel and other value-added by-products. This unique combination of fossil and renewables will provide a smooth transition for the critical energy supplies.

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Dr. Gan has successfully achieved several significant technological innovations at international advanced level with significant economic and social benefits. By innovating System Energy Efficiency technology, he pioneered "Ubiquitous Energy Network" by integrating energy and information networks and "Coal-based Near-zero CO<sub>2</sub> Emission Clean Energy Production System". His System Energy Efficiency technology was demonstrated through 2010 Shanghai World Expo "China Pavilion" and "Eco-home". His "Eco-city" development model has been widely recognized and applied to ten cities both at home and

abroad.



**Junjie Gu**, Professor at Carleton University (Canada), received engineering degrees from Tianjin University (China), the University of Kaiserslautern (Germany) and trained as postdoctoral fellow at the University of Toronto, and has been serving as a professor at Carleton University from 2002. During his sabbatical leave in China in 2008, served as the Chief Scientist of Coal Chemistry and Group Leader for "Large Scale Coal-based Clean Energy and Coal Gasification Technology", with ENN Science and Technology development Co., Ltd. (China). Dr. Gu is active in the area of Energy Technologies, Heat and Mass Transfer, Thermodynamics, especially on Coal Gasification, Supercritical Water Cooled Nuclear Reactors, Thermal Driven Refrigeration Systems, etc. He received various awards from organizations, including: ENN Merit Award (2009); Petro-Canada Young Innovator Award (2007); Research Achievement Award, Carleton University (2006); Recognition Award, SAE VTMS (2005); NSERC Postdoctoral Fellowship (2000-2002); DAAD (German Academic Exchange Service) Scholarship (1995-2000). He has filed over 40 patents worldwide, and published over 100 papers in the field of clean energy conversions.